08-03-00

PCT $ US

FORM PTO-1390 (REV 10-94) U.S. DEPARTMENT OF COMMERCE PATENT AND TRADEMARK OFFICE

TRANSMITTAL LETTER TO THE UNITED STATES DESIGNATED/ELECTED OFFICE (DO/EO/US) CONCERNING A FILING UNDER 35 U.S.C. 371

ATTORNEY'S DOCKET NUMBER	
00-434	
U.S. APPLICATION NO. (If known, see 37 C.F.R. 1.5)	09/601709

INTERNATIONAL APPLICATION NO.	INTERNATIONAL FILING DATE	PRIORITY DATE CLAIMED
PCT/CH99/00050	February 5, 1999	February 5, 1998

TITLE OF INVENTION
POLAR POLYMERIC COATING

APPLICANT(S) FOR DO/EO/US
EVA MARIA MOSER

Applicant herewith submits to the United States Designated/Elected Office (DO/EO/US) the following items and other information:

1. XX This is a **FIRST** submission of items concerning a filing under 35 U.S.C. 371.
2. ☐ This is a **SECOND or SUBSEQUENT** submission of items concerning a filing under 35 U.S.C. 371.
3. ☐ This express request to begin national examination procedures (35 U.S.C. 371(f)) at any time rather than delay examination until the expiration of the applicable time limit set in 35 U.S.C. 371(b) and PCT Articles 22 and 39(1).
4. XX A proper Demand for International Preliminary Examination was made by the 19th month from the earliest claimed priority date.
5. ☐ A copy of the International Application as filed (35 U.S.C. 371(c)(2))
   a. ☐ is transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ has been transmitted by the International Bureau.
   c. ☐ is not required, as the application was filed in the United States Receiving Office (RO/US)
6. XX A translation of the International Application into English (35 U.S.C. 371(c)(2)).
7. ☐ Amendments to the claims of the International Application under PCT Article 19 (35 U.S.C. 371(c)(3))
   a. ☐ are transmitted herewith (required only if not transmitted by the International Bureau).
   b. ☐ have been transmitted by the International Bureau.
   c. ☐ have not been made; however, the time limit for making such amendments has NOT expired.
   d. ☐ have not been made and will not be made.
8. ☐ A translation of the amendments to the claims under PCT Article 19 (35 U.S.C. 371(c)(3)).
9. XX An oath or declaration of the inventor(s) (35 U.S.C. 371(c)(4)).
10. XX A translation of the annexes to the International Preliminary Examination Report under PCT Article 36 (35 U.S.C. 371(c)(5)). Amended Sheets 1, 2, 3, 3A, 7, 10, 11 and 12

Items 11. to 16. below concern document(s) or information included:

11. ☐ An Information Disclosure Statement under 37 CFR 1.97 and 1.98.
12. XX An assignment document for recording. A separate cover sheet in compliance with 37 CFR 3.28 and 3.31 is included.
13. XX A FIRST preliminary amendment.
    ☐ A SECOND or SUBSEQUENT preliminary amendment.
14. ☐ A substitute specification.
15. ☐ A change of power of attorney and/or address letter.
16. ☐ Other items or information:

(January 1995)

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on August 2, 2000
(Date of Deposit)

Antoinette Sullo
Name and Reg. No. of Attorney

Antoinette Sullo
Signature

8-2-00
Date of Signature

U.S. APPLICATION NO. (if known, see 37 CFR 1.5)	INTERNATIONAL APPLICATION NO.	ATTORNEY'S DOCKET NUMBER
09/601709	PCT/CH99/00050	00-434

17. [XX] The following fees are submitted:

	CALCULATIONS PTO USE ONLY	
BASIC NATIONAL FEE (37 CFR 1.492(a)(1)-(5)):		
Search Report has been prepared by the EPO or JPO..... $840		
International preliminary examination fee paid to USPTO (37 CFR 1.482) $660.00		
No international preliminary examination fee paid to USPTO (37 CFR 1.482) but international search fee paid to USPTO (37 CFR 1.445(a)(2))..$730.00		
Neither international preliminary examination fee (37 CFR 1.482) nor international search fee (37 CFR 1.445(a)(2)) paid to USPTO..... $980.00		
International preliminary examination fee paid to USPTO (37 CFR 1.482) and all claims satisfied provisions of PCT Article 33(2)-(4)..... $92.00		
ENTER APPROPRIATE BASIC FEE AMOUNT =	$ 840.00	
Surcharge of $130.00 for furnishing the oath or declaration later than [ ]20 [ ]30 months from the earliest claimed priority date (37 CFR 1.492(e)). See Prel. Amdt.	$	

CLAIMS	NUMBER FILED		NUMBER EXTRA	RATE		
Total claims	20	-20 =		X $22.00	$	
Independent claims	1	-3 =		X $76.00	$	
MULTIPLE DEPENDENT CLAIM(S) (if applicable)				+ $240.00	$	
TOTAL OF ABOVE CALCULATIONS =					$ 840.00	
Reduction by 1/2 for filing by small entity, if applicable. Verified Small Entity Statement must also be filed (Note 37 CFR 1.9, 1.27, 1.28).					$ --	
SUBTOTAL =					$ 840.00	
Processing fee of $130.00 for furnishing the English translation later than [ ] 20 [ ] 30 months from the earliest claimed priority date (37 CFR 1.492(f)). +					$ --	
TOTAL NATIONAL FEE =					$ 840.00	
Fee for recording the enclosed assignment (37 CFR 1.21(h)). The assignment must be accompanied by an appropriate cover sheet (37 CFR 3.28, 3.31). $40.00 per property +					$ 40.00	
TOTAL FEES ENCLOSED =					$ 880.00	
					Amount to be: refunded	$
					charged	$

a. [X] A check in the amount of $ 880.00 to cover the above fees is enclosed.

b. [ ] Please charge my Deposit Account No. _____ in the amount of $_____ to cover the above fees. A duplicate copy of this sheet is enclosed.

c. [X] The Commissioner is hereby authorized to charge any additional fees which may be required, or credit any overpayment to Deposit Account No. 02-0184. A duplicate copy of this sheet is enclosed.

NOTE: Where an appropriate time limit under 37 CFR 1.494 or 1.495 has not been met, a petition to revive (37 CFR 1.137(a) or (b)) must be filed and granted to restore the application to pending status.

SIGNATURE

Robert H. Bachman
NAME

19,374
REGISTRATION NUMBER

SEND ALL CORRESPONDENCE TO:

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Form PTO-1390 (REV 10-94) page 2 of 2

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant : EVA MARIA MOSER Docket No.: 00-434

Serial No.: Examiner :

Filed : Art Unit :

For : POLAR POLYMERIC COATING

900 Chapel Street
Suite 1201
New Haven, CT 06510-2802

PRELIMINARY AMENDMENT

Hon. Commissioner of Patents and Trademarks
United States Patent and Trademark office
Washington, D.C. 20231

Dear Sir:

In the above-identified application for United States patent, please amend as follows:

IN THE SPECIFICATION

Page 3

(Amended Page), line 14, "carbonic acids" should read --carboxylic acid--.

Page 4

line 17, "polyamide" should read --polyimide--.

Page 6

line 28, "better" should read --lower--.

Page 7

(Amended Sheet), line 19, "nM/m" should read --mN/m--.

Page 8

line 2, "in" should read --and--.

IN THE CLAIMS

Prior to calculating the filing fee due, please amend the following claims.

In claim 3, line 1, delete "any of claims 1 or 2" and insert --claim 1--.

In claim 4, line 1, delete "any of claims 1 to 3" and insert --claim 1--.

In claim 5, line 1, delete "any of claims 1 to 4" and insert --claim 1--; and on line 4, delete "carbonic acid" and insert --carboxylic acid--.

In claim 6, line 1, delete "any of claims 1 to 5" and insert --claim 1--.

In claim 7, line 1, delete "any of claims 1 to 6" and insert --claim 1--.

In claim 17, line 2, delete "any of claims 1 to 7" and insert --claim 1--.

In claim 18, line 1, delete "any of claims 1 to 16" and insert --claim 1--.

In claim 19, line 1, delete "any of claims 1 to 16" and insert --claim 1--.

In claim 20, line 1, delete "any of claims 1 to 16" and insert --claim 1--.

REMARKS

The instant Preliminary Amendment is submitted so as to correct errors appearing in the translation and to delete the multiple dependent claims and therefore save on filing costs.

An early action on the merits is requested.

Respectfully submitted,

EVA MARIA MOSER

By _____
Robert H. Bachman
Attorney for Applicant

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on August 2, 2000
(Date of Deposit)

Antoinette Sullo
Name and Reg. No. of Attorney

Antoinette Sullo
Signature

8-2-00
Date of Signature

Express Mail Label No. EL398542749US

Date: August 2, 2000

WO99/39842 (as amended)

Polar Polymer-Like Coating

The present invention concerns a process for coating of substrates by means of plasma polymerisation. The invention also concerns a coating, produced using the process, of a polymer substrate and applications of the process.

Polymer substrates such as in particular flexible substrates are coated amongst other reasons in order to influence the surface composition or appearance of the polymer or protect the surface mechanically, physically and chemically. This may be to increase the adhesion to the surface or the printability, to prepare the surface for further functional coatings, to ensure protection against abrasion or damage, to reduce or prevent the permeability of certain gases or liquids on or through the surface of the substrate, or to increase the chemical resistance of the substrate to certain chemicals.

For surface treatment of polymer substrates which increases the polarity or surface tension in the short term, a multiplicity of methods are known where in principle two processes occur most commonly: modification of the surface for example by a corona discharge at atmospheric pressure or by a plasma process at reduced pressure.

Both said processes are important in particular in connection with the increase in adhesion to the polymer substrate or the increase in printability. However, in corona discharge it has been found that the printability for example of polymer packing films is good only immediately after performance of the treatment and the printability diminishes again after just a few hours or days.

In contrast, in a series of documents it is proposed to modify or coat the polymer by means of a low pressure plasma process, where the coating is usually hydrophilic and allows

good adhesion or printability. This printability is retained practically without restriction because of the coating.

Thus for example in JP-59-15569 and WO,A1 AU89/00220 it is proposed to coat a polymer substrate by means of plasma polymerisation of an organic compound, together for example with a working gas and water or water vapour. It is also proposed in WO95/04609 to treat or coat the surface by means of plasma polymerisation of an organic compound in the presence of hydrogen peroxide.

US,A 3397132 concerns a coating of metal surfaces, where an electric discharge occurs in the presence of organic gases and an inert carrier gas. With regard to the inorganic gases, the absence of water is neither mentioned nor otherwise stated as essential. In contrast, precise statements are made for other parameters such as pressure, temperature, concentration, voltage and frequency. Corresponding modifications to the parameters achieve the desired improvements in the metal surfaces by plasma coating.

In a polar plasma coating to DE, A1 3908418, at least one organic compound and an optional inorganic gas is used. Plastic containers are coated on the inside with coatings impermeable to organic solvents, where the inside of the container is impacted with a low pressure plasma. This process too does not mention the absence of water.

Firstly, the coatings proposed in the state of the art have a poor adhesion to the substrate, or they have restricted wettability. The use of peroxide or water and oxygen causes a problem as the resulting "working gas" is aggressive and can attack the surface of the substrate (etching).

It is therefore a task of the present invention to propose a process for polar coating of substrates by means of plasma polymerisation which does not have the present

disadvantages. A coating produced with the process and applications are also proposed.

With reference to the process, the task according to the invention is solved with the features according to claim 1.

The hydrocarbon compounds which have up to a maximum of eight carbon atoms are therefore of relatively low molecular weight, so the compounds have a relatively high vapour pressure at room temperature.

Preferred substances are alkanes, alkenes, alkynes (acetylene), polyenes, monovalent or multivalent alcohols, carbonic acids, ethers, aldehydes and/or ketones. These can be aliphatic, cycloaliphatic or aromatic hydrocarbon compounds.

The use of water vapour as a process gas in a gas discharge is anything but ideal and must be avoided according to the invention. Furthermore, a water-containing layer would have a lower chemical and thermal resistance which would have negative effect on the subsequent process stages and the definition and stability of the coatings. The plasma-polymerised coating according to the invention is therefore water-free and so compact that although hydrophilic it absorbs almost no water in further processing.

For this reason in each case it is essential for the invention that the process gas used for plasma polymerisation or the working gas is free from water or water vapour. The absence of water or water vapour at least in the process gas in any case ensures that the working gas or gas mixture contains no peroxide compounds which could for example form in the plasma chamber if water and oxygen are used.

Merely by the simultaneous use of oxygen and hydrogen in the process gas, or oxygen- and hydrogen-containing compounds

such as for example ethanol or methanol, is it possible for water vapour or peroxide to form during the process, but only traces of these components which usually do not have a negative effect on the coating. Also the formation of water vapour or peroxide can be predicted and controlled and thus limited.

A comparison with the known coatings for example from the three said documents from the state of the art, shows such a high hydrophility of the coatings on the polymer substrate that a substantially better printability is achieved. This

is achieved even after storage of at least six months. It is assumed that this improvement in the properties of the coating proposed according to the invention is attributable to the circumstance that the process gas used in the process according to the invention is free from water or water vapour.

In principle all known plasma processes such as for example microwave discharge, high or low frequency discharge, DC magnetron discharge, arc vaporisation, the use of electron guns etc. are suitable for the performance of the process according to the invention. The process proposed according to the invention is also suitable for coating all known polymer substrates used today, for example for the production of packing materials such as polyethylene, polyamide, polypropylene, PMMA, PVC, polyesters such as PETP, PBTP, polyamide, polycarbonate etc. It is also possible to coat metal and ceramic substrates. The polar coating can then serve as a coupling agent between these materials and further coatings such as for example corrosion protection coatings, or allow the connection of different materials such as for example metal/polymer etc.

By means of the process proposed according to the invention, the said polymer substrate is given a polar polymer-like coating or a plasma coating with high surface tension in which are integrated polar groups such as for example hydroxyl, carboxyl, carbonyl groups (see figs 2a and 2b) or NO_x groups, whereby on the surface of this coating an excellent adhesion can be achieved for polar functional layers and/or polar materials, which is reflected for example in an excellent printability. In particular packaging materials, films, containers, bottles made from the said polymer substrates can thus be processed considerably more easily. Usually a coating of the order of a few nm is sufficient to achieve this increased adhesion and printability.

As already stated, for performance of the proposed process, all low pressure plasma processes known and commonly used today can be used, so detailed description of these processes can be omitted at this point. The substrate to be coated, flexible for example, such as a film, hollow body or similar, is placed in a vacuum chamber into which is introduced the working gas consisting of the said components. As already stated it is essential that this working gas is free from water or water vapour or moisture. Then by means of the plasma process a plasma-polymerised coating is deposited on the surface of the material to be coated.

It is also possible to coat a granulate or powder according to the invention and then produce a polar film or body from this (Ref. 2).

The coating thus generated by plasma-polymerisation usually has a layer thickness of a few nm, for example between 1 and 100, preferably 5 to 20 nm; but it can also amount to a few µm. Evidently the layer thickness depends on the requirements, whether in addition to the printability a scratch protection or anti-fog effect is required, to which the coating achieved according to the invention can also make a contribution.

Also the ratio between the inorganic gas components such as for example oxygen, nitrogen, ammonia or carbon monoxide or carbon dioxide, and the organic compound, depends on the properties required for the coating. The ratio can vary greatly depending on the components contained in the gas mixture or working gas. Table 1 compares two examples. In addition to the said components, naturally further constituents such as in particular inert gases for example argon or helium etc., can be used.

Suitable organic compounds are in particular alkanes with a chain length of up to around eight carbon atoms such as for

example methane, ethane, propane etc. Also alkenes such as ethylene, propylene etc. are suitable as organic compounds.

Also suitable are acetylenes or acetylene-based compounds such as so-called alkynes.

Equally suitable are polyenes, i.e. hydrocarbons with several double bonds, again with up to around eight carbon atoms.

Also suitable are alcohols such as methanol, ethanol, propanol etc. and multivalent alcohols such as for example ethylene glycol.

Also suitable are monovalent or multivalent organic acids, ethers, aldehydes and ketones. The hydrocarbon compounds stated can be aliphatic, cycloaliphatic or aromatic hydrocarbons, where naturally all the said compounds can also be substituted such as for example by amino groups, halogens, ammonia etc.

The present invention will now be explained in more detail using the examples below:

Examples: stable hydrophilic surfaces by plasma-polymerised functional coating with polar groups:

At a basic pressure of for example better than 3×10^{-6} mbar, a plasma reactor is flooded with the process gas mixture until the required process pressure is achieved, for example 1.6×10^{-2} mbar. In the present examples a microwave discharge (2.45 GHz) was then ignited while the process gases were supplied continuously. A coating with a polar proportion of 41% and a surface tension of 50 mN/m was achieved with a gas mixture of 48 sccm (standard cubic cm per minute) CO2, 12 sccm CH4 and 12 sccm Ar with a microwave power of 62 Watts (specimen 10/PET). The substrate was a 12 µm thin PET film or a 20 µm thin polypropylene film

(specimen 2/BOPP), representative of polymer substrates. An increase in process pressure up to atmospheric pressure leads to a high deposition rate and is presently the state of optimisation of coatings. Table 1 also shows that by varying the power and process gas mixture, the required surface tension for the corresponding substrate can be achieved. Comparison of the various gas mixtures in table 1 shows that the gas mixture has a greater influence on the hydrophility than varying the power supplied to the plasma by 80 Watts. Table 1 shows the coatings which were produced between July and October 1997 and for which the surface tension was again measured in January 1998 and 1999.

After 12 weeks, in no coating was a total surface tension of less than 45 mN/m measured, which is of decisive importance for the subsequent process stages in production. Specimen 1/PET was produced on 16th July 1997, where the surface tension after 6 months was still 47 mN/m and after 18 months 49 nM/m. In contrast, with corona treatment and surface modification with low pressure plasmas (with process gases containing oxygen and/or nitrogen), after a few weeks no such high surface tension was measured. According to literature the plasma-modified surface is restructured in the first three weeks following treatment (Ref. 1). As the stability of the hydrophilic layer was monitored for more than 18 months, it can safely be assumed that a stable state has been achieved as the surface tension and polarity values of the coatings after around two months were only insignificantly modified, as is shown for example from Fig 3.

The chemical structure of the hydrophilic layers is clear from the enclosed figures 2a and 2b. The two figures 2a and 2b show the XPS spectra (= X-ray photo-electron spectroscopy) of C (1s), specimens 8 and 10 (PET) on table 1. The surface areas shown in figures 2a and 2b are representative of the following bonds: 1 for O-C=O, 3 for C=O, 5 for C-O, 7 for C-H. C-O bonds are present in alcohol

and ether, C=O in ketones and aldehydes and O-C=O in esters in carboxylic acids.

In figure 2a the area proportion of 1 is 6.5%, the area proportion of 3 is 8.9%, the proportion of 5 is 20.1% and the proportion of 7 is 64.5%. The total proportion of carbon is 76.2% and that of oxygen 23.8%. The ratio of carbon to oxygen is therefore 76.2 : 23.8.

In figure 2b the area proportion of 1 is 15.4%, the area of 3 is 2.6%, the area of 5 is 20.0% and the area of 7 is 61.9%. The proportion of C (1s) is 70.0% and the proportion of O (1s) is 30.0%.

The XPS (X-ray photo-electron spectroscopy) results show that the polar surface of the specimen 10/PET in comparison with specimen 8/PET contains 6 at% more oxygen and this is present mainly in ester and carboxylic compounds. (Hydrogen cannot be detected with this method). In both specimens (8/PET and 10/PET) one-fifth of the oxygen is bonded as alcohol or ether. The higher polarity (polar proportion / total surface tension) of 41% (specimen 10/PET) in contrast to 33% (specimen 8/PET) is consequently due to a higher oxidation of the carbon atoms (O-C=O).

By means of the process described above as an example, a series of PET and BOPP films were coated, the total surface tension and polarity of the coatings of which were then determined. The coating parameters and results of the measurements are summarised in the table 1 below.

PET: Polyethylene terephthalate film 20 µm thick
BOPP: Biaxial-oriented polypropylene 20 µm thick

The wettability of all samples or coatings listed in table 1 is between 20 and 63 mN/m (to DIN-EN 828 (draft)). In relation to the examples of generated coatings summarised in table 1, it is important to emphasise that the coatings

generated in this way remain polar. As has been proven, these remain polar for at least twelve months from which it can presumably be concluded that these coatings remain stable for years.

The test conditions described as examples above serve merely to explain in more detail the basic concept of the present invention. Naturally it is also possible to produce plasma-polymerised coatings according to the process defined in the invention under widely varying conditions and on very different substrates. The coating (any functional coating which is polar in nature), printing, laminating (adhesion - gluing to polar adhesives) is possible on such a polar surface for new printing agents and adhesives based on the solvent water. In order to stabilise the surface tension, doping of the coating with inorganic anions (nitrogen, fluorine etc.) and inorganic cations (metals or metal oxides) is also permitted. Thus further properties, e.g. the electrical conductivity of the coating, can be adjusted as required for the product.

It is essential for the invention that the working gas used for plasma polymerisation is free from water and water vapour and moisture.

(Ref. 1): Thomas R. Gengenbach et al., "Concurrent Restructuring and Oxidation of the Surface of n-Hexane Plasma Polymers During Ageing in Air", Plasmas and Polymers, Vol. 1, No. 3, 1996, p. 207 - 228.

(Ref. 2): J. Messelhäuser, S. Berger, "Plasma Modification of Powdery Plastics", 7th Federal German Seminar, 13th - 14th March 1996, Rub-Bochum, p. 39 ff.

CLAIMS

1. Process for coating substrates with a polar coating by means of plasma polymerisation,
characterised in that
to produce a coating whic is stable in the long term, a water-free process gas is used which contains at least one substituted hydrocarbon compound with up to a maximum of eight C-atoms and also an inorganic gas.

2. Process according to claim 1, characterised in that the proportion of organic compound in the gas mixture is between 5 and 90 volume %.

3. Process according to any of claims 1 or 2, characterised in that the inorganic gas is oxygen, a halogen, hydrogen, an inert gas, carbon monoxide, carbon dioxide, nitrogen and/or another nitrogen-containing gas.

4. Process according to any of claims 1 to 3, characterised in that an aliphatic, aliphatic cyclic and/or aromatic hydrocarbon is used.

5. Process according to any of claims 1 to 4, characterised in that as an organic compound a polyene, a monovalent or multivalent alcohol, a monovalent or multivalent carbonic acid, ether, aldehyde and/or a ketone is used.

6. Process according to any of claims 1 to 5, characterised in that as an organic compound
   - an alkane such as methane, ethane, propane, butane, pentane and/or hexane,
   - an alkene such as ethylene, butylene, propylene and/or isopropylene, or
   - an alkyne such as acetylene or a derivative of acetylene is used.

7. Process according to any of claims 1 to 6, characterised in that a fluorine-, nitrogen- or sulphur-substituted hydrocarbon compound is used as an organic compound.

8. Process according to claim 1, characterised in that a substrate is coated with two to four gases of the groups consisting of CO_2, CH_4, O_2, C_2H_2, NH_3 and Ar.

9. Process according to claim 8, characterised in that a substrate is coated with a process gas of CO_2, C_2H_2 and Ar, preferably in the volume ratio of 4:1:1.

10. Process according to claim 8, characterised in that a substrate is coated with a process gas of NH_3, CO_2, CH_4 and Ar, preferably in the volume ratio of 2:1:1:1 or 2:2:1:1.

11. Process according to claim 8, characterised in that a substrate is coated with a process gas of CO_2 and CH_4, preferably in the volume ratio of 2:1 or 4:1.

12. Process according to claim 8, characterised in that a substrate is coated with a process gas of CO_2, CH_4 and Ar, preferably in the volume ratio of 3:3:1 or 4:1:1.

13. Process according to claim 8, characterised in that a substrate is coated with a process gas of CO_2 and Ar, preferably in the volume ratio of 4:1.

14. Process according to claim 8, characterised in that a substrate is coated with a process gas of CH_4, O_2 and Ar, preferably in the volume ratio of 1:1:1.

15. Process according to claim 8, characterised in that a substrate is coated with a process gas of CO_2, CH_4, O_2

and Ar, preferably in the volume ratio of 1:2:1:2 or 1:4:1:2.

16. Process according to claim 8, characterised in that a substrate is coated with a process gas of CH_4, NH_3 and Ar, preferably in the volume ratio of 2:2:1, 1:4:1 or 1:2:1.

17. Polymer coating of a substrate produced by means of a process according to any of claims 1 to 7, characterised in that the polar coating has an initial surface tension of < 45 mN/m which remains unchanged for at least one year.

18. Use of the process according to any of claims 1 to 16 for coating polymer flexible substrates, polymer substrates reinforced with ceramic fibres, glass fibres, polymer fibres and/or carbon fibres, and powder- or granulate-formed substrates for production of a polar film or a polar moulded body.

19. Use of the process according to any of claims 1 to 16 for coating packing materials such as in particular films, bottles and other containers, and substrates for adhesion of composite materials.

20. Use of the process according to any of claims 1 to 16 for coating ceramic or metal substrates.

Polar Polymer-like Plasma Coating

Sample/Film	Power [W]	CO_2 [sccm]	CH_4 [sccm]	Ar [sccm]	O_2 [sccm]	C_2H_2 [sccm]	NH_3 [sccm]	Thickness [nm]	Surface Tension. [mN/m] January 1998	Surface Tension. [mN/m] January 1999	Polarity [%] Jan. 99	Polarity [%] Jan. 99
1/PET	95	48	-	12	-	-	-	26	47	49	30	38
2/PET	105	48	24	-	-	-	-	32	45	49	33	33
3/PET	126	48	24	-	-	-	-	30	47	47	30	35
4/PET	88	48	24	12	-	-	-	24	48	47	32	31
5/PET	78	36	36	12	-	-	-	80	45	44	27	27
6/PET	99	48	12	-	-	-	-	10	50	48	39	34
7/PET	75	48	24	-	-	-	-	45	49	47	33	32
8/PET	123	48	24	-	-	-	-	64	**48**	**45**	**33**	**33**
9/PET	137	36	36	12	-	-	-	168	45	44	26	29
10/PET	62	48	12	12	-	-	-	9	**52**	**50**	**41**	**39**
11/PET	103	48	12	12	-	-	-	11	50	48	40	40
12/PET	106	48	-	12	-	12	-	284	**51**	**52**	**29**	**36**
13/PET	100	-	12	12	12	-	-	18	51	50	39	38
14/PET	110	12	-	12	6	-	-	12	47	45	32	33
15/PET	104	6	12	12	6	-	-	8	49	46	34	34
16/PET	108	6	24	12	6	-	-	22	45	45	28	30
1/BOPP	70	48	12	12	-	-	-	10	52	52	42	42
2/BOPP	60	48	12	12	-	-	-	11	51	51	42	42
17/PET	100	-	12	12	-	-	48	27	-	58	-	60
18/PET	250	-	12	12	-	-	24	35	-	53	-	55
19/PET	200	-	24	12	-	-	24	46	-	45	-	41
20/PET	120	24	12	12	-	-	24	25	-	54	-	43
21/PET	130	12	12	12	-	-	24	18	-	**63**	-	**65**
22/PET	115	24	12	12	-	-	24	16	-	56	-	50
12 µm PET-Film									42.0		7	
20 µm BOPP-Film									30.2		3	

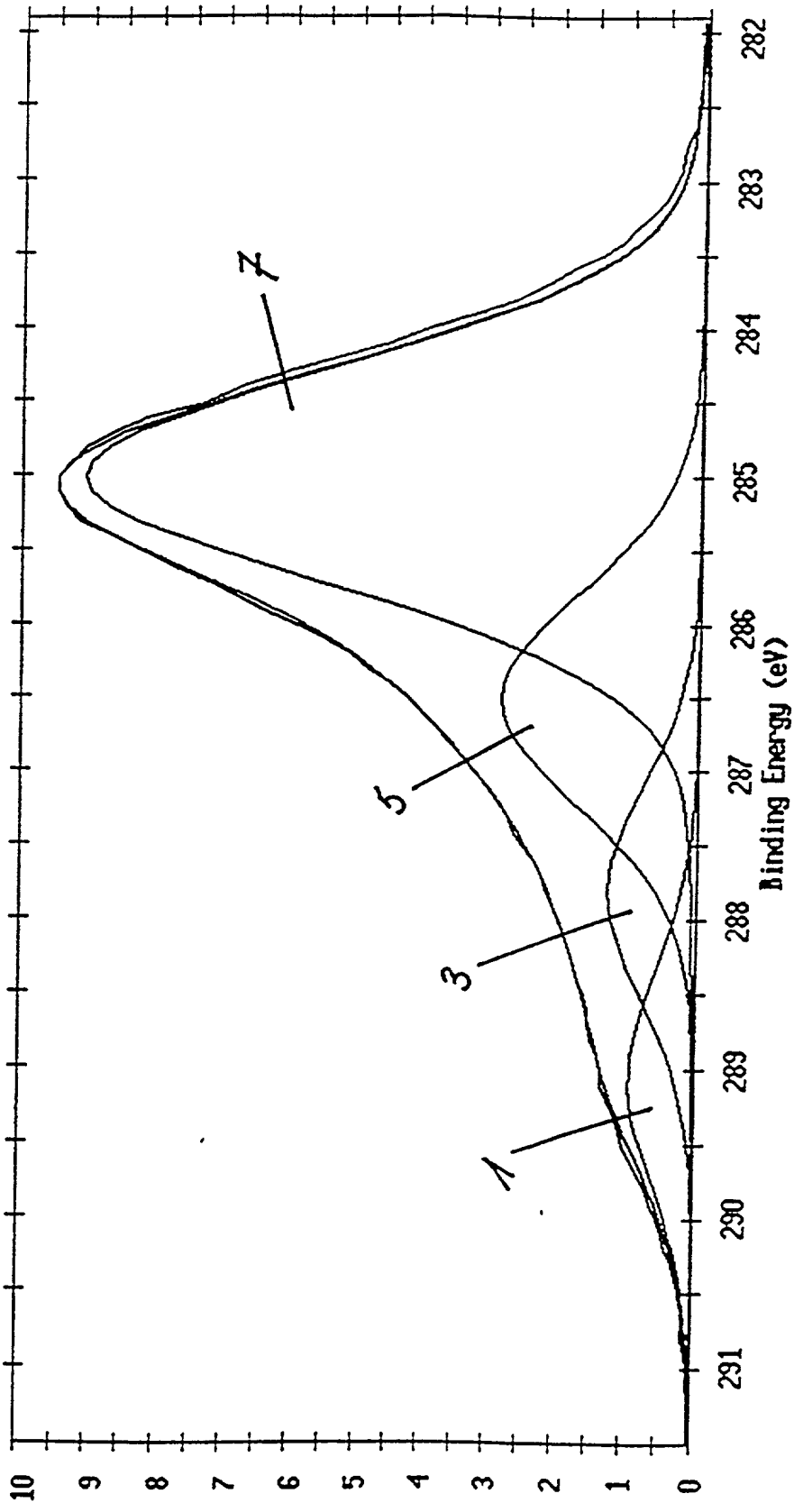

N(E)/E, SHF
Binding Energy (eV)
0
1
2
3
4
5
6
7
8
9
10
282
283
284
285
286
287
288
289
290
291
1
2
3
5

Fig. 2a

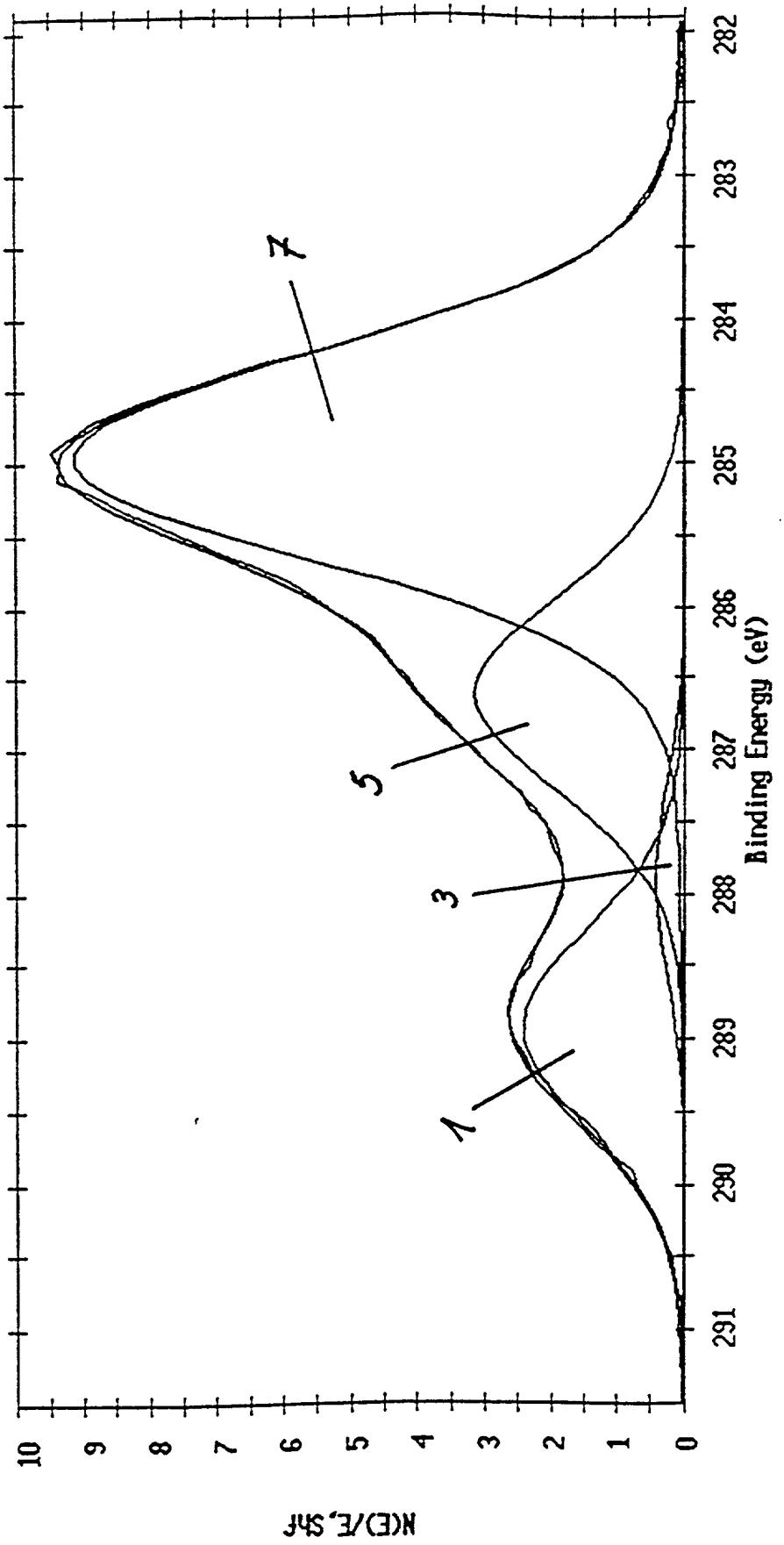
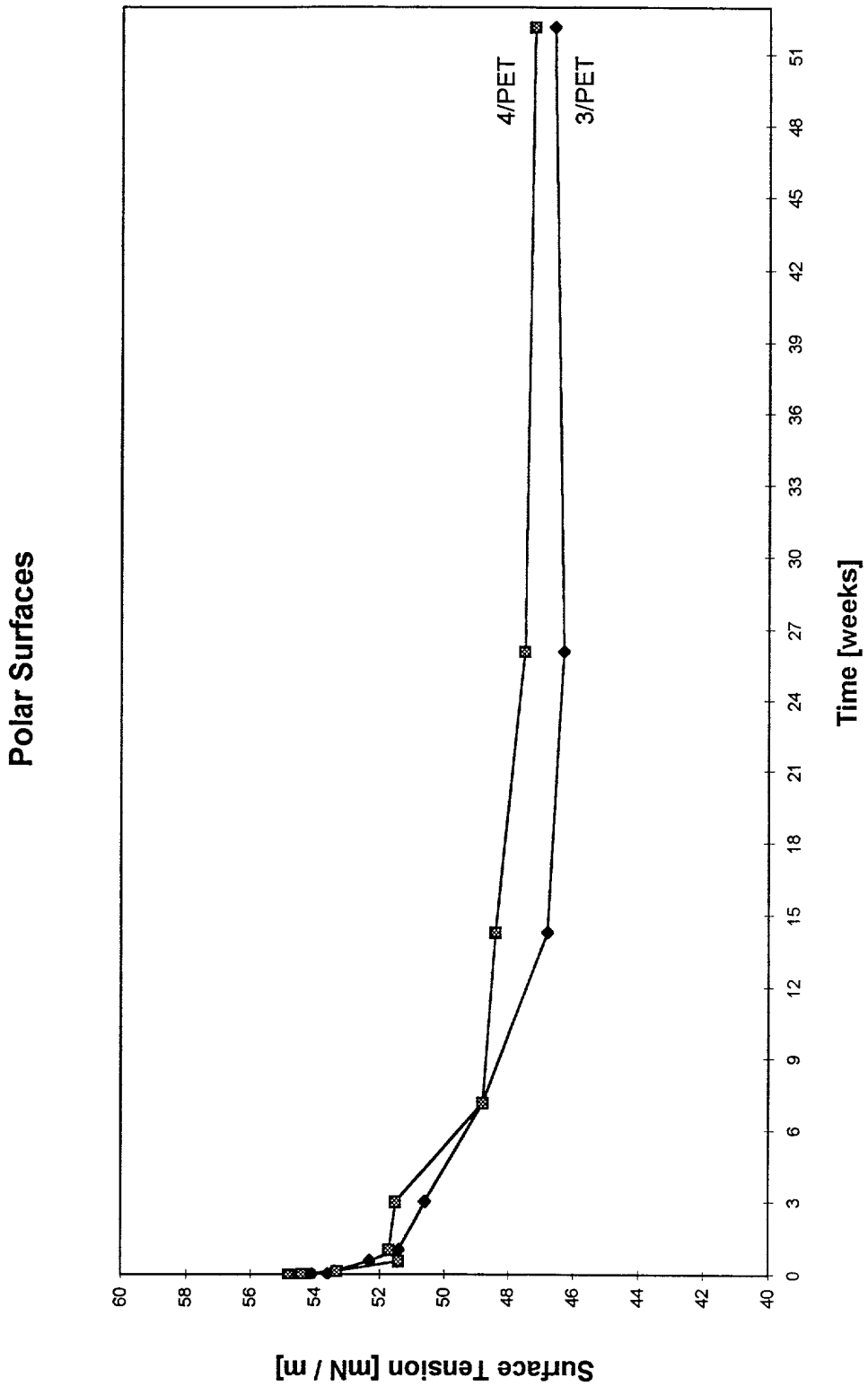

N(E)/E, shf
10
9
8
7
6
5
4
3
2
1
0
282
283
284
285
286
287
288
289
290
291
Binding Energy (eV)
7
5
3
1

Fig. 2b


Polar Surfaces
Surface Tension [mN / m]
Time [weeks]
4/PET
3/PET
60
58
56
54
52
50
48
46
44
42
40
0
3
6
9
12
15
18
21
24
27
30
33
36
39
42
45
48
51

Practitioner's Docket No. 00-434 ***PATENT***

COMBINED DECLARATION AND POWER OF ATTORNEY

(ORIGINAL, DESIGN, NATIONAL STAGE OF PCT, SUPPLEMENTAL, DIVISIONAL, CONTINUATION, OR C-I-P)

As a below named inventor, I hereby declare that:

TYPE OF DECLARATION

This declaration is of the following type:

(check one applicable item below)

☐ original.

☐ design.

NOTE: With the exception of a supplemental oath or declaration submitted in a reissue, a supplemental oath or declaration is not treated as an amendment under 37 CFR 1.312 (Amendments after allowance). M.P.E.P. § 714.16, 7th Edition.

☐ supplemental.

NOTE: If the declaration is for an International Application being filed as a divisional, continuation or continuation-in-part application, do not check next item; check appropriate one of last three items.

XX national stage of PCT.

NOTE: If one of the following 3 items apply, then complete and also attach ADDED PAGES FOR DIVISIONAL, CONTINUATION OR C-I-P.

NOTE: See 37 C.F.R. § 1.63(d) (continued prosecution application) for use of a prior nonprovisional application declaration in the continuation or divisional application being filed on behalf of the same or fewer of the inventors named in the prior application.

☐ divisional.

☐ continuation.

NOTE: Where an application discloses and claims subject matter not disclosed in the prior application, or a continuation or divisional application names an inventor not named in the prior application, a continuation-in-part application must be filed under 37 C.F.R. § 1.53(b) (application filing requirements — nonprovisional application).

☐ continuation-in-part (C-I-P).

INVENTORSHIP IDENTIFICATION

WARNING: *If the inventors are each not the inventors of all the claims, an explanation of the facts, including the ownership of all the claims at the time the last claimed invention was made, should be submitted.*

My residence, post office address and citizenship are as stated below, next to my name. I believe that I am the original, first and sole inventor *(if only one name is listed below)* or an original, first and joint inventor *(if plural names are listed below)* of the subject matter that is claimed, and for which a patent is sought on the invention entitled:

TITLE OF INVENTION

POLAR POLYMERIC COATING

SPECIFICATION IDENTIFICATION

the specification of which:

(complete (a), (b), or (c))

(a) ☐ is attached hereto.

NOTE: "The following combinations of information supplied in an oath or declaration filed on the application filing date with a specification are acceptable as minimums for identifying a specification and compliance with any one of the items below will be accepted as complying with the identification requirement of 37 CFR 1.63:

"(1) name of inventor(s), and reference to an attached specification which is both attached to the oath or declaration at the time of execution and submitted with the oath or declaration on filing;

"(2) name of inventor(s), and attorney docket number which was on the specification as filed; or

"(3) name of inventor(s), and title which was on the specification as filed."

Notice of July 13, 1995 (1177 O.G. 60).

(b) ☐ was filed on _____, as ☐ Serial No. 0 /_____ or ☐ _____ and was amended on _____ *(if applicable).*

NOTE: Amendments filed after the original papers are deposited with the PTO that contain new matter are not accorded a filing date by being referred to in the declaration. Accordingly, the amendments involved are those filed with the application papers or, in the case of a supplemental declaration, are those amendments claiming matter not encompassed in the original statement of invention or claims. See 37 C.F.R. § 1.67.

NOTE: "The following combinations of information supplied in an oath or declaration filed after the filing date are acceptable as minimums for identifying a specification and compliance with any one of the items below will be accepted as complying with the identification requirement of 37 CFR 1.63:

"(A) application number (consisting of the series code and the serial number, e.g., 08/123,456);

"(B) serial number and filing date;

"(C) attorney docket number which was on the specification as filed;

"(D) title which was on the specification as filed and reference to an attached specification which is both attached to the oath or declaration at the time of execution and submitted with the oath or declaration; or

"(E) title which was on the specification as filed and accompanied by a cover letter accurately identifying the application for which it was intended by either the application number (consisting of the series code and the serial number, e.g., 08/123,456), or serial number and filing date. Absent any statement(s) to the contrary, it will be presumed that the application filed in the PTO is the application which the inventor(s) executed by signing the oath or declaration."

M.P.E.P. § 601.01(a), 7th Ed.

(c) ☒ was described and claimed in PCT International Application No. PCT/CH99/00050, filed on February 5, 1999 and as amended under PCT Article 19 on _____ *(if any).*

SUPPLEMENTAL DECLARATION (37 C.F.R. § 1.67(b))

(complete the following where a supplemental declaration is being submitted)

☐ I hereby declare that the subject matter of the

☐ attached amendment

☐ amendment filed on _____

was part of my/our invention and was invented before the filing date of the original application, above-identified, for such invention.

ACKNOWLEDGEMENT OF REVIEW OF PAPERS AND DUTY OF CANDOR

I hereby state that I have reviewed and understand the contents of the above-identified specification, including the claims, as amended by any amendment referred to above.

I acknowledge the duty to disclose information, which is material to patentability as defined in 37, Code of Federal Regulations, § 1.56,

(also check the following items, if desired)

☒ and which is material to the examination of this application, namely, information where there is a substantial likelihood that a reasonable Examiner would consider it important in deciding whether to allow the application to issue as a patent, and

☐ in compliance with this duty, there is attached an information disclosure statement, in accordance with 37 C.F.R. § 1.98.

PRIORITY CLAIM (35 U.S.C. §§ 119(a)–(d))

NOTE: *"The claim to priority need be in no special form and may be made by the attorney or agent if the foreign application is referred to in the oath or declaration as required by § 1.63. The claim for priority and the certified copy of the foreign application specified in 35 U.S.C. 119(b) must be filed in the case of an interference (§ 1.630), when necessary to overcome the date of a reference relied upon by the examiner, when specifically required by the examiner, and in all other situations, before the patent is granted. If the claim for priority or the certified copy of the foreign application is filed after the date the issue fee is paid, it must be accompanied by a petition requesting entry and by the fee set forth in § 1.17(i). If the certified copy is not in the English language, a translation need not be filed except in the case of interference; or when necessary to overcome the date of a reference relied upon by the examiner; or when specifically required by the examiner, in which event an English language translation must be filed together with a statement that the translation of the certified copy is accurate." 37 C.F.R. § 1.55(a).*

I hereby claim foreign priority benefits under Title 35, United States Code, §§ 119(a)–(d) of any foreign application(s) for patent or inventor's certificate or of any PCT international application(s) designating at least one country other than the United States of America listed below and have also identified below any foreign application(s) for patent or inventor's certificate or any PCT international application(s) designating at least one country other than the United States of America filed by me on the same subject matter having a filing date before that of the application(s) of which priority is claimed.

(complete (d) or (e))

(d) ☐ no such applications have been filed.

(e) ☒ such applications have been filed as follows.

NOTE: *Where item (c) is entered above and the International Application which designated the U.S. itself claimed priority check item (e), enter the details below and make the priority claim.*

PRIOR FOREIGN/PCT APPLICATION(S) FILED WITHIN 12 MONTHS (6 MONTHS FOR DESIGN) PRIOR TO THIS APPLICATION AND ANY PRIORITY CLAIMS UNDER 35 U.S.C. § 119(a)-(d)

COUNTRY (OR INDICATE IF PCT)	APPLICATION NUMBER	DATE OF FILING (day, month, year)	PRIORITY CLAIMED UNDER 37 USC 119
Switzerland	281/98	05/02/98	☒ YES NO ☐
			☐ YES NO ☐
			☐ YES NO ☐
			☐ YES NO ☐
			☐ YES NO ☐

CLAIM FOR BENEFIT OF PRIOR U.S. PROVISIONAL APPLICATION(S)
(34 U.S.C. § 119(e))

I hereby claim the benefit under Title 35, United States Code, § 119(e) of any United States provisional application(s) listed below:

PROVISIONAL APPLICATION NUMBER	FILING DATE
_____ /_____	_____
_____ /_____	_____
_____ /_____	_____

CLAIM FOR BENEFIT OF EARLIER US/PCT APPLICATION(S) UNDER 35 U.S.C. § 120

☐ The claim for the benefit of any such applications are set forth in the attached ADDED PAGES TO COMBINED DECLARATION AND POWER OF ATTORNEY FOR DIVISIONAL, CONTINUATION OR CONTINUATION-IN PART (C-I-P) APPLICATION.

ALL FOREIGN APPLICATION(S), *IF ANY*, FILED MORE THAN 12 MONTHS (6 MONTHS FOR DESIGN) PRIOR TO THIS U.S. APPLICATION

NOTE: *If the application filed more than 12 months from the filing date of this application is a PCT filing forming the basis for this application entering the United States as (1) the national stage, or (2) a continuation, divisional, or continuation-in-part, then also complete ADDED PAGES TO COMBINED DECLARATION AND POWER OF ATTORNEY FOR DIVISIONAL, CONTINUATION OR C-I-P APPLICATION for benefit of the prior U.S. or PCT application(s) under 35 U.S.C. § 120.*

POWER OF ATTORNEY

I hereby appoint the following practitioner(s) to prosecute this application and transact all business in the Patent and Trademark Office connected therewith.

(list name and registration number)

(4) Robert H. Bachman (19,374), Gregory P. LaPointe (28,395), Barry L. Kelmachter (29,999) and George A. Coury (34,309)

(check the following item, if applicable)

☐ I hereby appoint the practitioner(s) associated with the Customer Number provided below to prosecute this application and to transact all business in the Patent and Trademark Office connected therewith.

☐ Attached, as part of this declaration and power of attorney, is the authorization of the above-named practitioner(s) to accept and follow instructions from my representative(s).

NOTE: *"Special care should be taken in continuation or divisional applications to ensure that any change of correspondence address in a prior application is reflected in the continuation or divisional application. For example, where a copy of the oath or declaration from the prior application is submitted for a continuation or divisional application filed under 37 CFR 1.53(b) and the copy of the oath or declaration from the prior application designates an old correspondence address, the Office may not recognize, in the continuation or divisional application, the change of correspondence address made during the prosecution of the prior application. Applicant is required to identify the change of correspondence address in the continuation or divisional application to ensure that communications from the Office are mailed to the current correspondence address. 37 CFR 1.63(d)(4)." § 601.03, M.P.E.P., 7th Edition.*

SEND CORRESPONDENCE TO	DIRECT TELEPHONE CALLS TO: *(Name and telephone number)*
☒ Address Bachman & LaPointe, P.C. 900 Chapel Street, Suite 1201 New Haven, CT 06510-2802	Robert H. Bachman (203) 777-6628
☐ Customer Number _____	

(complete the following if applicable)

Since this filing is a ☐ continuation ☐ divisional there is attached hereto a Change of Correspondence Address so that there will be no question as to where the PTO should direct all correspondence.

DECLARATION

I hereby declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

SIGNATURE(S)

NOTE: *Carefully indicate the family (or last) name, as it should appear on the filing receipt and all other documents.*

NOTE: *Each inventor must be identified by full name, including the family name, and at least one given name without abbreviation together with any other given name or initial, and by his/her residence, post office address and country of citizenship. 37 CFR § 1.63(a)(3).*

NOTE: *Inventors may execute separate declarations/oaths provided each declaration/oath sets forth all the inventors. Section 1.63(a)(3) requires that a declaration/oath, inter alia, identify each inventor and prohibits the execution of separate declarations/oaths which each sets forth only the name of the executing inventor. 62 Fed. Reg. 53,131, 53,142, October 10, 1997,*

1-00

Full name of sole or first inventor

EVA	MARIA	MOSER
(GIVEN NAME)	*(MIDDLE INITIAL OR NAME)*	*FAMILY (OR LAST NAME)*

Inventor's signature Eva Maria Moser

Date 10-07-2000 **Country of Citizenship** SWITZERLAND

Residence Gütschstrasse 32, CH-8122 Binz, Switzerland CHX

Post Office Address SAME AS ABOVE

Full name of second joint inventor, if any

(GIVEN NAME)	*(MIDDLE INITIAL OR NAME)*	*FAMILY (OR LAST NAME)*

Inventor's signature __________

Date __________ **Country of Citizenship** __________

Residence __________

Post Office Address __________

Full name of third joint inventor, if any

(GIVEN NAME)	*(MIDDLE INITIAL OR NAME)*	*FAMILY (OR LAST NAME)*

Inventor's signature __________

Date __________ **Country of Citizenship** __________

Residence __________

Post Office Address __________

(check proper box(es) for any of the following added page(s) that form a part of this declaration)

☐ **Signature** for fourth and subsequent joint inventors. *Number of pages added* _____.

* * *

☐ **Signature** by administrator(trix), executor(trix) or legal representative for deceased or incapacitated inventor. *Number of pages added* _____.

* * *

☐ **Signature** for inventor who refuses to sign or cannot be reached by person authorized under 37 CFR 1.47. *Number of pages added* _____.

* * *

☐ Added page for **signature** by one joint inventor on behalf of deceased inventor(s) where legal representative cannot be appointed in time. (37 CFR 1.47)

* * *

☐ Added pages to combined declaration and power of attorney for divisional, continuation, or continuation-in-part (C-I-P) application.

☐ Number of pages added _____

* * *

☐ Authorization of practitioner(s) to accept and follow instructions from representative.

* * *

(if no further pages form a part of this Declaration, then end this Declaration with this page and check the following item)

☒ This declaration ends with this page.